Takaki Laboratory (Special Issue on the Commemoration of the Fortieth Anniversary)

TAKAKI LABORATORY (December 1951~)

Head: Dr. Hideo Takaki

After Professor H. Hirata's passing away in December, 1950, many projections of investigations on the inner structure of metals and alloys and other subjects were passed to the 2nd Laboratory of Professor Dr. Senji Utzino, the then director of the institute. That is, studies were made of both the oriented structure of ionic crystals by growth and plastic deformation and the epitaxy growth of metals deposited on mica and others by Masaaki Yanagisawa[1,4]. Studies were also made of the effects of co-existed ions on the inner structure of copper deposited on zinc surface from cupric sulphate solution by Katsuyuki Yasuda and Hidekiyo Fujihira[5]. Further studies were made of the inner structure change due to rolling and annealing in aluminium by Masashige Koyama[2,3].

In December, 1951, the 2nd Laboratory of Prof. S. Utzino was closed and a new laboratory was established for Dr. Hideo Takaki, Professor of Metal Physics in the Departement of Chemistry, Faculty of Science, Kyoto University and excellent studies were developed on the following chief subjects.

I. Crystal Growth and Substructures of Metals and Alloys

Single crystals of ferromagnetic materials, nickel and silicon steel, were produced by Bridgman method from the melt by Takaki and his co-workers[13,15,16]. In that case, it was observed that the rate of crystal growth was related to the crystallographic orientation and the crystals grown were composed of a bundle of rod-like sub-crystals with a few degrees of misorientation each other. It has been well known that the so-called columnar structure in cast-metals and cast-alloys is related to the crystal structure. The crystallographic orientation of columnar crystals, for example, is parallel to [001] in cubic metals. Therefore, in the light of anisotropy of the rate of crystal growth, an X-ray examination was carried out by Takaki and Koyama on the cast structure of an ingot of 4 % silicon steel (930 kg in weight) produced at Kawasaki Iron Mfg. Co. (bottom poured)[8]. Further, minute X-ray examinations were performed on high purity aluminium ingots (99.993 %) cast by an unidirectional cooling method[12,16]. In the ingot solidified at a rapid rate of cooling, was observed the [001] arrangement of columnar crystals which was resulted from the surviving of favourably oriented crystals. Even the unfavourably oriented crystals, however, survived at a slow rate of cooling. A line structure was observed in the columnar crystals mentioned above. The slower the rate of cooling, the spacing between the lines was wider. The misorientation between both the neighbouring parts of lines was half degree to one. The line structure had already been observed on the preparation of tin single crystals by Takaki, Koyama and

Fujihira[9]. About that time, two types of substructures in metal crystals grown from the melt were published by B. Chalmers and his co-workers: one was the corrugated substructure originating in the solute impurities; the other was the striated one bounded by the low angle tilt-subboundaries and a mechanism of collapsing vacancy discs was suggested for the generation of edge dislocations.

Under impetus from the studies of Chalmers et al. mentioned above, further minute studies on crystal growth, substructures and production of perfect crystals were also proceeded in the Laboratory with tin, lead, zinc and nickel as follows.

On an autoradiographic examination of nickel crystals using the isotope S^{35} as an impurity tracer, it was confirmed by Takaki, Shigeo Maeda and Yasuto Nakai that the corrugated substructure was caused by the impurity[24]. Further a type of pre-dendritic growth form called "partial dendrite" by Takaki, Koyama and Fujihira was found in tin at the transition of growth type from cellular (corresponding to the section of corrugations) to cellular dendritic[18,19,20,22]. A transition of growth form from free dendritic to cellular dendritic and a further transition from cellular dendritic to cellular were observed by Yoshikazu Tsujii, Maeda, Koyama and Takaki in lead crystals grown from the seed crystal at relatively rapid lowering rates[59]. Maeda measured a change of electric resistance as the temperature falling in a molten thread of tin about 0.1 mm in diameter and a relation between the rate of dendritic growth and the degree of supercooling in tin was discussed from the result obtained.

X-ray studies on perfection of tin single crystals grown from the melt and studies on production of perfect crystals of tin were made by Koyama using X-ray diffraction microscopy and transmission type bent-quartz monochromater[25,26,30,31,34]. A technique to prevent the transmission of subboundaries from the seed was also devised by Maeda in which the transmission of pre-existing striated subboundaries was suppressed by crystal growing through a glass tube tip inserted into an outer glass tube[23].

A study on nickel single crystals was carried out by Maeda to discuss the relation of the spacing between subboundaries to the growth rate which was introduced by F.C. Frank[35]. Further, for one reason of the investigation of a relationship between the thickness and the formation of striated subboundaries in the crystals grown from the melt, and for the other reason of the direct observation of dislocations in thin crystals by Lang's X-ray diffraction method, a technique for producing single crystal foils of tin from the melt was devised by Takaki, Koyama, Tsujii, Kōzō Iwauchi and Maeda using a special type of glass mould. Single crystal foils of tin about 70 μ in thickness were actually produced by the technique mentioned above[43,53]. Various improvements, however, have now been attempted to prevent an effect of stress induced by an adhesion between the crystal surface and the inner wall of glass mould.

The so-called facet growth pattern was observed by Maeda on the top free surface of rod-like single crystals of nickel. Planes of (111), (100) and (110) were also assumed as the growth planes under a consideration that the surface energy is roughly proportional to the product of the surface atomic density times the number

of missing bonds per atom on the surface. It was found by Iwauchi, Tsujii, Maeda, Koyama and Takaki that the preferred orientation of rod-like zinc crystals grown from the melt (99.999%) was parallel to the basal plane. The preference, further, was explained by the growth of the grains developed from the nuclei satisfying the minimum surface free energy in the heterogeneous nucleation on the inner wall of a glass crucible[54].

As a different category from that concerning the crystal growth and substructures in metals and alloys mentioned above, the following studies have been made by Part-time Instructor, Apollo Saika†.

II. Studies in High-Resolution Nuclear Magnetic Resonance

They are concerned mainly with theoretical aspects of chemical shift and nuclear spin-spin coupling and are now being proceeded in the Department of Chemistry, Faculty of Science, Kyoto University.

The experimental results obtained in the Laboratory during fifteen years have been published in the following papers.

Pubications

(* indicates an article published in Japanese)

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2. M. Koyama: Rolling and Annealing of Aluminium, *ibid.*, **28,** 41 (1952).*
3. M. Koyama: X-Ray Studies on the Inner Structural Change Due to the Annealing in Aluminium, *ibid.*, **28,** 58 (1952).
4. M. Yanagisawa: Epitaxy Growth of Metals Deposited on Mica, Calcite, Iceland Spar, Rock Salt and Glass, *ibid.*, **28,** 58 (1952).
5. H. Fujihira: Effects of Co-existed Ions on the Inner Structure of Copper Deposited on Zinc Surface from Cupric Sulphate Solution, *ibid.*, **28,** 59 (1952).
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